

MAY. 10. 2006 4:04PM

NO. 1365 P. 4/9

Application No.: 10/530,226

Docket No.: 12834-00011-US

AMENDMENTS TO THE CLAIMS

1. (Original) A proton-conducting polymer membrane which comprises polyazole blends and is obtainable by a process comprising the steps
  - A) preparation of a mixture comprising polyphosphoric acid, at least one polyazole (polymer A) and/or one or more compounds which are suitable for forming polyazoles under the action of heat according to step B),
  - B) heating of the mixture obtainable according to step A) under inert gas to temperatures of up to 400°C,
  - C) application of a layer using the mixture from step A) and/or B) to a support,
  - D) treatment of the membrane formed in step C) until it is self-supporting,wherein at least one further polymer (polymer B) which is not a polyazole is added to the composition obtainable according to step A) and/or step B) and the weight ratio of polyazole to polymer B is in the range from 0.1 to 50.
2. (Original) The membrane as claimed in claim 1, characterized in that the mixture prepared in step A) comprises compounds which are suitable for forming polyazoles under the action of heat in step B), with these compounds comprising one or more aromatic and/or heteroaromatic tetraamino compounds and one or more aromatic and/or heteroaromatic carboxylic acids or derivatives thereof which have at least two acid groups per carboxylic acid monomer and/or one or more aromatic and/or heteroaromatic diaminocarboxylic acids.
3. (Original) The membrane as claimed in claim 1, characterized in that the mixture prepared in step A) comprises compounds which are suitable for forming polyazoles under the action of heat in step B), with these compounds being obtainable by reaction of one or more aromatic and/or heteroaromatic tetraamino compounds with one or more aromatic and/or heteroaromatic carboxylic acids or derivatives thereof which have at least two acid groups per carboxylic acid monomer or of one or more aromatic and/or heteroaromatic diaminocarboxylic

463735

2

MAY. 10. 2006 4:04PM

NO. 1365 P. 5/9

Application No.: 10/530,226

Docket No.: 12834-00011-US

acids in the melt at temperatures of up to 400°C.

4. (Previously presented) The membrane as claimed in claim 2, characterized in that aromatic and/or heteroaromatic tetraamino compounds used as compounds suitable for forming polyazoles comprise compounds selected from the group consisting of 3,3',4,4'-tetraaminobiphenyl, 2,3,5,6-tetraaminopyridine and 1,2,4,5-tetraaminobenzene.

5. (Previously presented) The membrane as claimed in claim 2, characterized in that aromatic and/or heteroaromatic carboxylic acids or derivatives thereof having at least two acid groups per carboxylic acid monomer used as compounds suitable for forming polyazoles comprise compounds selected from the group consisting of isophthalic acid, terephthalic acid, phthalic acid, 5-hydroxyisophthalic acid, 4-hydroxyisophthalic acid, 2-hydroxyterephthalic acid, 5-aminoisophthalic acid, 5-N,N-dimethylaminoisophthalic acid, 5-N,N-diethylaminoisophthalic acid, 2,5-dihydroxyterephthalic acid, 2,5-dihydroxyisophthalic acid, 2,3-dihydroxyisophthalic acid, 2,3-dihydroxyphthalic acid, 2,4-dihydroxyphthalic acid, 3,4-dihydroxyphthalic acid, 3-fluorophthalic acid, 5-fluoroisophthalic acid, 2-fluoroterephthalic acid, tetrafluorophthalic acid, tetrafluoroisophthalic acid, tetrafluoroterephthalic acid, 1,4-naphthalenedicarboxylic acid, 1,5-naphthalenedicarboxylic acid, 2,6-naphthalenedicarboxylic acid, 2,7-naphthalenedicarboxylic acid, diphenic acid, 1,8-dihydroxynaphthalene-3,6-dicarboxylic acid, bis(4-carboxyphenyl) ether, benzophenone-4,4'-dicarboxylic acid, bis(4-dicarboxyphenyl) sulfone, biphenyl-4,4'-dicarboxylic acid, 4-trifluoromethylphthalic acid, 2,2-bis(4-carboxyphenyl)hexafluoropropane, 4,4'-stilbenedicarboxylic acid, 4-carboxycinnamic acid, their C1-C20-alkyl esters, their C5-C12-aryl esters, their acid anhydrides and their acid chlorides.

6. (Previously presented) The membrane as claimed in claim 2 characterized in that the compounds suitable for forming polyazoles comprise aromatic tricarboxylic acids, their C1-C20-alkyl esters or C5-C12-aryl esters or their acid anhydrides or their acid halides or tetracarboxylic acids, their C1-C20-alkyl esters or C5-C12-aryl esters or their acid anhydrides or their acid halides.

7. (Original) The membrane as claimed in claim 6, characterized in that the aromatic tricarboxylic acids comprise compounds selected from the group consisting of 1,3,5-benzenetricarboxylic acid (trimesic acid); 2,4,5-benzenetricarboxylic acid (trimellitic acid); (2-

463735

3

AGE 5/9 \* RCVD AT 5/10/2006 4:02:55 PM [Eastern Daylight Time] \* SVR:USPTO-EFXXF-1/16 \* DNIS:2738300 \* CSID:302 661 2331 \* DURATION (mm:ss):02:42

MAY. 10. 2006 4:04PM

NO. 1365 P. 6/9

Application No.: 10/530,226

Docket No.: 12834-00011-US

carboxyphenyl)iminodiacetic acid, 3,5,3'-biphenyltricarboxylic acid; 3,5,4'-biphenyltricarboxylic acid, 2,4,6-pyridinetricarboxylic acid, benzene-1,2,4,5-tetracarboxylic acid; naphthalene-1,4,5,8-tetracarboxylic acid, 3,5,3',5'-biphenyltetracarboxylic acid, benzophenonetetracarboxylic acid, 3,3',4,4'-biphenyltetracarboxylic acid, 2,2',3,3'-biphenyltetracarboxylic acid, 1,2,5,6-naphthalenetetracarboxylic acid and 1,4,5,8-naphthalenetetracarboxylic acid.

8. (Previously presented) The membrane as claimed claim 6, characterized in that the content of tricarboxylic acid and/or tetracarboxylic acids is from 0 to 30 mol based on dicarboxylic acid used.

9. (Previously presented) The membrane as claimed in claim 2, characterized in that the compounds suitable for forming polyazoles comprise heteroaromatic dicarboxylic acids, tricarboxylic acids and/or tetracarboxylic acids which contain at least one nitrogen, oxygen, sulfur or phosphorus atom in the aromatics.

10. (Original) The membrane as claimed in claim 9, characterized in that pyridine-2,5-dicarboxylic acid, pyridine-3,5-dicarboxylic acid, pyridine-2,6-dicarboxylic acid, pyridine-2,4-dicarboxylic acid, 4-phenyl-2,5-pyridinedicarboxylic acid, 3,5-pyrazoledicarboxylic acid, 2,6-pyrimidinedicarboxylic acid, 2,5-pyrazinedicarboxylic acid, 2,4,6-pyridinetricarboxylic acid, benzimidazole-5,6-dicarboxylic acid, and also their C1-C20-alkyl esters or C5-C12-aryl esters, or their acid anhydrides or their acid chlorides are used.

11. (Previously presented) The membrane as claimed in claim 2, characterized in that the compounds suitable for forming polyazoles comprise diaminobenzoic acid and/or its monohydrochloride and dihydrochloride derivatives.

12. (Original) The membrane as claimed in claim 1, characterized in that the polymer B) is used in step A) in an amount in the range from 10 to 50% by weight, based on the weight of the mixture A) and/or B).

13. (Original) The membrane as claimed in claim 1, characterized in that the polymer B) comprises at least one polyolefin.

14. (Original) The membrane as claimed in claim 1, characterized in that the polymer B) comprises at least one polymer having C-O bonds.

463735

4

PAGE 6/9 \* RCVD AT 5/10/2006 4:02:55 PM [Eastern Daylight Time] \* SVR:USPTO-EFXXF-1/16 \* DNIS:2738300 \* CSID:302 661 2331 \* DURATION (mm:ss):02:42

MAY. 10. 2006 4:04PM

NO. 1365 P. 7/9

Application No.: 10/530,226

Docket No.: 12834-00011-US

15. (Original) The membrane as claimed in claim 1, characterized in that the polymer B) comprises at least one polymer having C-S bonds.
16. (Original) The membrane as claimed in claim 1, characterized in that the polymer B) comprises at least one polymer having C-N bonds.
17. (Original) The membrane as claimed in claim 1, characterized in that the polymer B) comprises at least one inorganic polymer.
18. (Original) The membrane as claimed in claim 1, characterized in that the polymer B) comprises at least one sulfonated polymer.
19. (Currently amended) The membrane as claimed in claim 1, characterized in that the heating according to step B) is carried out after the formation of a sheet-like structure according to step C).
20. (Original) The membrane as claimed in claim 1, characterized in that the treatment according to step D) is carried out at temperatures in the range from 0°C to 150°C in the presence of moisture.
21. (Original) The membrane as claimed in claim 1, characterized in that the treatment of the membrane in step D) is carried out for from 10 seconds to 300 hours.
22. (Original) The membrane as claimed in claim 1, characterized in that the membrane formed after step D) is crosslinked by action of oxygen.
23. (Original) The membrane as claimed in claim 1, characterized in that a layer having a thickness of from 20 to 4000  $\mu\text{m}$  is produced in step C).
24. (Original) The membrane as claimed in claim 1, characterized in that the membrane formed after step D) has a thickness of from 15 to 3000  $\mu\text{m}$ .
25. (Original) An electrode provided with a proton-conducting polymer coating which comprises polyazole blends and is obtained by a process comprising the steps
- A) preparation of a mixture comprising  
polyphosphoric acid,

MAY. 10. 2006 4:05PM

NO. 1365 P. 8/9

Application No.: 10/530,226

Docket No.: 12834-00011-US

- at least one polyazole (polymer A) and/or one or more compounds which are suitable for forming polyazoles under the action of heat according to step B),
- B) heating of the mixture obtainable according to step A) under inert gas to temperatures of up to 400°C,
- C) application of a layer using the mixture from step A) and/or B) to an electrode,
- D) treatment of the membrane formed in step C),

wherein at least one further polymer (polymer B) which is not a polyazole is added to the composition obtainable according to step A) and/or step B) and the weight ratio of polyazole to polymer B is in the range from 0.1 to 50.

26. (Original) The electrode as claimed in claim 24, wherein the coating has a thickness of from 2 to 3000 µm.
27. (Previously presented) A membrane-electrode unit comprising at least one electrode and at least one membrane as claimed in claim 1.
28. (Currently amended) A membrane-electrode unit comprising ~~at least one electrode as claimed in claim 25 or 26 and~~ at least one membrane as claimed in claim 1.
29. (Previously presented) A fuel cell comprising one or more membrane-electrode units as claimed claim 27.
30. (New) A membrane-electrode unit comprising at least one electrode as claimed in claim 25.
31. (New) A membrane-electrode unit comprising at least one electrode as claimed in claim 26.

463735

6

PAGE 8/9 \* RCVD AT 5/10/2006 4:02:55 PM [Eastern Daylight Time] \* SVR:USPTO-EFAXF-1/16 \* DNIS:2738300 \* CSID:302 661 2331 \* DURATION (mm:ss):02:42